Modelling of groundwater contamination caused by lignite mining in Eastern Germany

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ABSTRACT

Open cast mining of lignite leads to dump sites containing high amounts of acidity. This is caused by weathering of reduced sulphur compounds of the tertiary coal concomitant layers Acid Mine Drainage (AMD). The „conveying bridge" technology commonly used in the East German open casts produce dump sites which are heterogeneous in detail but in a larger hydrogeological scale a homogenous mixture of the coal hanging layers. Therefore it is difficult to investigate such an artificial sediment body by point-shaped exposure. It is necessary to establish a balance of the dump with the chemical knowledge of the forefield sediments. For the prediction of the acidity potential the genetical understanding of the iron disulphide content within the different tertiary layers is decisive. For the coal concomitant layers of 2nd Lausatian coal seam an areal new data interpretation of multitude analysis from the seventies and eighties was performed. The results confirm the basic ideas of sedimentary pyrite formation by Berner [1984].

Mineralised dump waters migrating into the unworked surroundings are subjected to homogeneous and heterogeneous interactions. To model these processes successfully a reactive transport model is necessary. Such coupled models solve sequentially advective-dispersive transport equations and the resulting system of geochemical equations. The requirement of CPU- time for this procedure is very high. For that reason the use of the „Front Limitation" (FL) algorithm for solving the advective- dispersive transport equation is very advantageous. It overcomes the numerical restriction of grid size (Peclet criterion) and hence, reduces dimension of the model.

INTRODUCTION

Open cast mining of lignite leads to dumps containing high amounts of acidity. This is caused by the reduced sulphur compounds within the tertiary coal concomitant layers which are exposed to oxygen during coal output and inside the water unsatured dumps.

In the former GDR domestic lignite was the most important energy source. There are two large regions which are affected by the phenomena of AMD. These are the „Central German brown coal district " around Leipzig and the „Lower Lausatian brown coal district" near the border to Poland (Fig.1).
The majority of the open casts was closed down due to the drastic reduction of coal output since 1990. When redevelopment of steady state flow conditions in the dumps is finished there will be a migration of dump waters into the unworked surroundings. To evaluate the affect and danger of dump waters for the surrounding areas (e.g. waterworks, lakes) as well as the necessity and suitability of remedial measures, a predictive modelling of the transport processes is necessary. Therefore understanding of the reduced sulphur compound content of the different stratigraphic units as well as the buffering potential of the quaternary layers is essential.

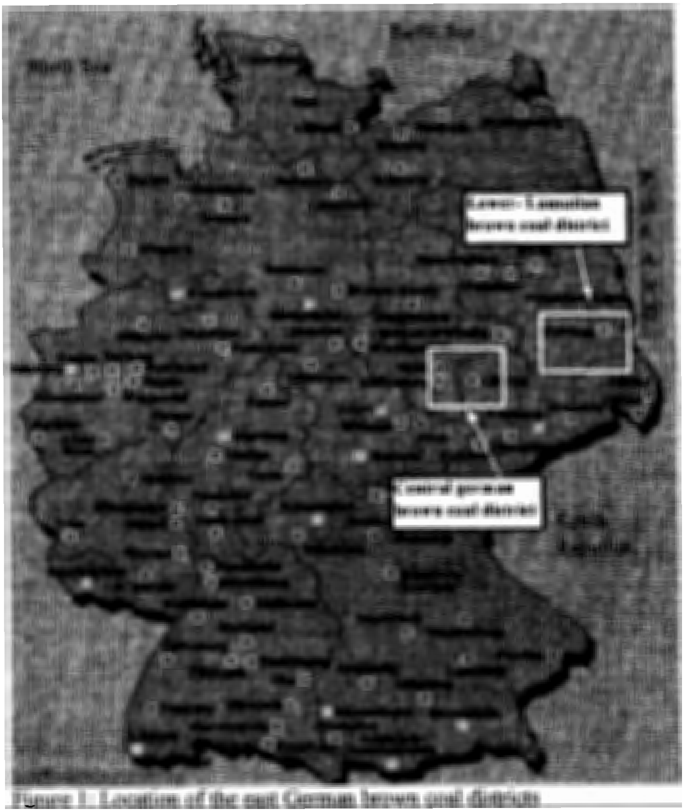


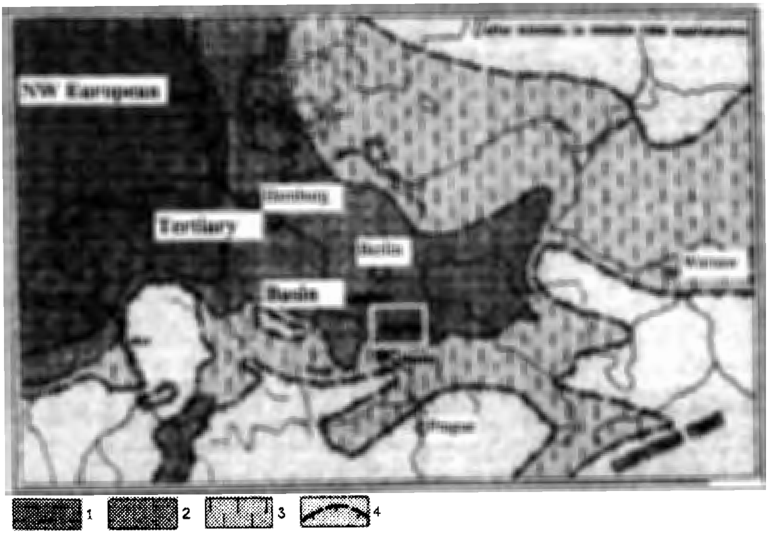
Figure 1: Location of the east German brown coal districts

GEOLOGICAL DEPOSIT CONDITIONS & MINING TECHNOLOGY

The two coal districts differ in their geological conditions. The following paragraphs describe the geological conditions and mining technology of the Lower Lausatian brown coal district.

Geological deposit conditions

The tertiary strata series of Lower Lausatian has an thickness of ca. 200 m. These are mostly Miocene sediments. The series includes 4 seam horizons and is build up very cyclical. The regular sequences of coal, silt to clay layers and sand results from the palaeogeographical location of these district. Figure 2 explain the marginal position of the area in the NW European Tertiary Basin during Lower /Middle Miocene. The paralic formed coal seams was brackish/marine influenced by varying intensity. The tertiary series is glacial superimposed. This will be expressed by glacial channels which have partly eroded the tertiary layers. The quaternary channel structures divide the extensive seams into coal fields and determine with this the proportions of several open casts. The normal thickness of quaternary series apart from the channel structures varying between 20 and 50 m. These are glaciofluviatil sands, boulder clay and glacilimnic sediments. The coal output is adjusted to the 2^{nd} Miocene seam horizon which is deposited 50 -70 m under floor.



1- marine 2- brackish/terrestrial 3- terrestrial 4- area of denudation

Figure 2: Palaeogeography Lower/ Middle Miocene - northern part of Central Europe (after Kockel in Vinken [1988])

Mining technology

In the East German lignite open casts the overburden was mostly removed by „conveying bridge" technology. Quaternary and tertiary coal hanging sediments are simultaneous cut from highwall and reach the dump by a large band conveyor, which spans the whole open cast. Therefore dump bodies are in detail heterogeneous but in an larger, hydrogeological scale a structured mixture of the whole coal hanging sediments. Kaubisch [1986] investigated mixing factors for this technology and showed values up to 0,9 (1 means ideal mixing).

The investigation of such artificial, mixing bodies by point-shaped exposure is very accidental and therefore difficult. It needs as an background information a „chemical balancing" of the forefield sediments. Therefore detailed information about thickness and content of relevant acidity and buffering potentials of the different stratigraphic units were combined. For that purpose it is necessary to understand distribution laws of reduced sulphur compounds within the tertiary layers.

DISTRIBUTION LAWS OF THE SULPHUR COMPOUNDS

Theory

Sedimentary pyrite formation is a synsedimentary to early diagenetic process and depends on facies conditions. Basic parts of process was summarised by BERNER [1984] (Fig.3). Under strong reduced conditions proceed sulphate reduction by availability of organic matter. Formed H_2S reacts with reactive iron over intermediate stages to thermodynamically more stable irondisulphide (modification pyrite for marine, non- acidic conditions).

In dependence of facial conditions one of the three items bold framed in figure 3 will be limiting factor of the whole process. Organic matter limits pyrite formation for marine deposition. This results in a strict correlation of organic carbon content (C_{org}) to sulphur (S_t) in this sediments. For data sets used by Berner [1984] ratios of C_{org} / S_t are mostly between 1 and 3. In contrast pyrite formation in non marine freshwater sediments is limited by sulphate. The sulphur content for this sediments is much lower and C_{org} / S_t ratios are considerably higher than 10 (data sets of Berner [1984]). Third case are euxinic conditions (limitation by reactive iron). It has to be noted that Berner used the total sulphur content (S_t) under assumption that the predominant part will be reduced sulphur. Vairavamurthy et.al. [1995] discuss the whole cycle of formation and transformation of sedimentary sulphur in much more detail. They show that formation of reduced inorganic and organic sulphur compounds are strongly connected. Hence, for a basic investigation of stratigraphic units it is justified to use S_t under assumption that sediments are out of sulphate.

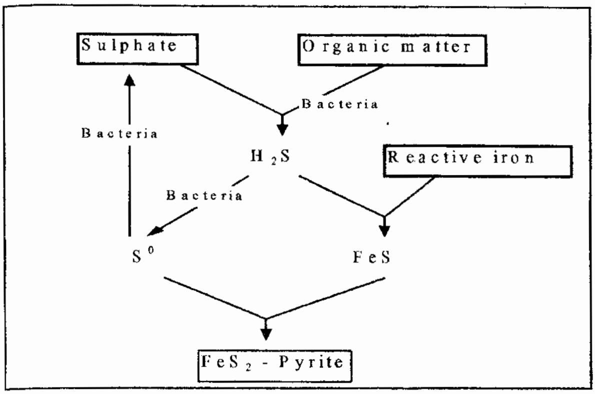


Figure 3: Model of pyrite formation (after BERNER [1984])

Data

Within the exploration of East German coal fields during the seventies and eighties a geochemical investigation of overburden sediments was systematically performed. Among the analysed parameters were: total sulphur content (S_t), total carbon content (C_t), cation exchange capacity (CEC), $CaCO_3$ content and a soil development by 10% hydrochloric acid (e.g. Frobenius [1977],.[1984], [1986]). For the TIC (total inorganic carbon) free tertiary layers the C_t values are equal to C_{org} .

Until now the interpretation of obtained data where not areal, not related to theory of sedimentary pyrite formation and much more with a lithological than a stratigraphical view.

Results

Derived from basic rules of sedimentary pyrite formation after Berner, C_{org} /S_t ratios for the different tertiary stratigraphic units had to be investigate. A areal data interpretation shows:

C_{org} / S_t ratios for stratigraphic units with different palaeogeographical positions confirm ideas after Berner. Figure 4 presents results for a part of the brackish/marine „overlying silt layers" of 2nd seam horizon. The C_{org} /S_t values vary between 3 and 5 by high sulphur content. Contrary to this are the „underlying silt layers". Figure 5 shows values for this stratigraphic unit which are higher than 20. The sulphur content in this probable terrestrial layers is much lower.

By means of the C_{org} /S_t ratios can be concluded that the reduced sulphur content distribution within the different tertiary units follows the basical ideas after Berner. In the different sequences of the Lausatian tertiary strata series the Transgressive Stand Track Sediments (compare Haq et.al. [1987] and Standke & Suhr et.al [1992]) include the highest S_{red} content. However the investigations of Rolland et.al. [1998] shows that sandy High Stand Track Sediments have lower content of S_{red} but acidification is much more important caused by higher diffusion coefficients than the silty Transgressive Stand Track Sediments. It is to summarise that the areal data interpretation together with modern analyses at the highwall and kinetically investigations there and within the unsaturated dumps (e.g. Rolland et.al. [1998]) allows to predict acidity potential of the overburden.

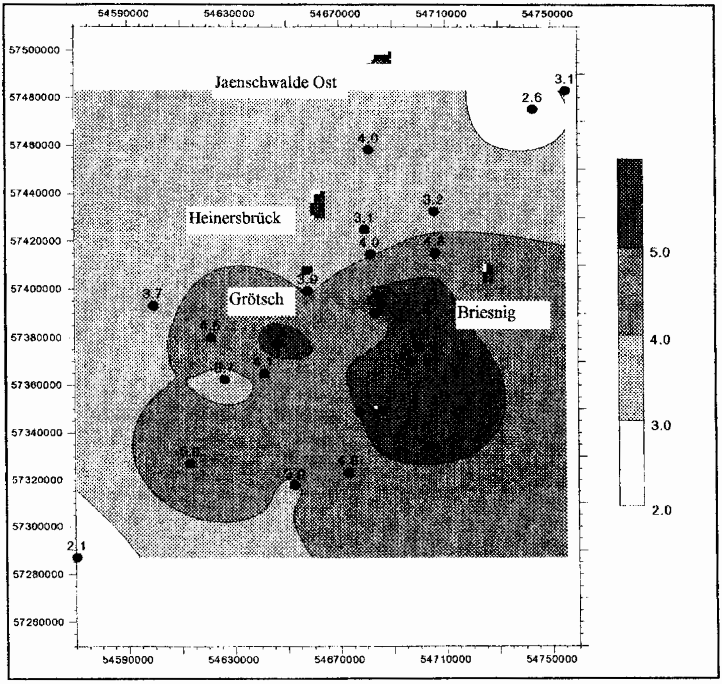


Figure 4 C_{org}/ S_t ratio of the „Overlying silt Layers“ of the 2nd Laustian Miocene Seam for a part of the „Lower Laustian coal district“ (data after FROBENIUS [1977,1984,1986])

TRANSPORT MODELLING

To estimate the groundwater conservation near dump sites it is necessary to model the migration process of developed dump waters into unworked surroundings. This process is characterised by homogeneous (redoxprocesses) and heterogeneous (mineral dissolution and precipitation, cation exchange) interactions. Transport models with isothermic approach are not able to cover complexity of the processes. Equation (1) shows that they are single component approaches and chemical interactions will only be included by fit parameters (retardation [R], rate constant of degradation [λ]).

$$div\left(D \cdot grad\, c - \frac{w}{n} \cdot c\right) - \lambda \cdot c \cdot R = R \cdot \frac{\partial c}{\partial t} - q \qquad (1)$$

Closer to reality is to model the process by a reactive transport model.

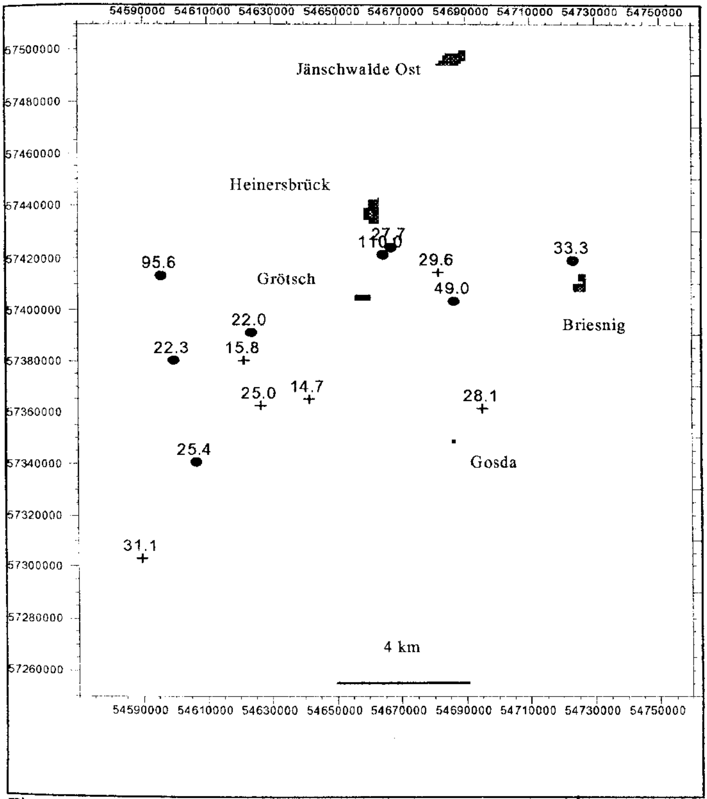


Figure 5: C_{org}/ S_t ratio of the „Underlying silt Layers" of the 2nd Laustian Miocene Seam for a part of the „Lower Laustian coal district" (data after FROBENIUS [1977,1984,1986])

Reactive transport model

The transport code PCGEOFIM® (Sames [1997]) was coupled with the geochemical equilibrium model PHREEQC (Parkhurst [1995]). The geochemical reactions are related to the transport equation by source-/sinkterm q_k in equation (2). Within PHREEQC the set of geochemical equations (represent the interactions) will be solved based on a thermodynamically database. The coupling procedure is similar to models like MINTRAN (Walter et.al.[1994]).

$$div\left(D \cdot grad\, c_k - \frac{w}{n} \cdot c_k\right) + q_k = \frac{\partial c_k}{\partial t} \qquad (2)$$

For time step t transport equation (2) is solved for N_x Cells and k master components. Sequentially simulation of chemical interactions at each cell within PHREEQC follows. The resulting changes of the $N_x \bullet k$ concentrations will be considered in the next time step t+1 within the transport equation as source-/sinkterm q_k.

It is evident that reactive transport models are much more complex than transport models with isothermic approach. This makes data supply much more difficult and increases CPU time requirement. Data supply means for example that it is necessary to characterise content of relevant secondary minerals in quantity (limits of X ray difractometry). The high CPU time requirement results from grid Peclet- and Courant number criterion.

Front Limitation algorithm

Up to now, the numerical solution of large transport problems requires an excessive amount of computer time. Finite Difference (FD), Control Volume (CV) and Finite Element (FE) algorithms have to take into account grid Peclet number and Courant number limits. The Peclet number must be lower than 2...10 to avoid numerical dispersion (Kinzelbach & Rausch, 1995), so that the maximum cell increment Δx is limited absolutely. If we suppose a longitudinal dispersivity δ_L in the order of magnitude of 10 m the maximum space increment in the horizontal direction has to be less than 20 ...100 m.

The Courant number has to be ≤ 1 to fulfil the local mass balance in each cell. At the end we have to realise that the both limits determine the space and time increments. The computer time expense (E) necessary can be estimated for a simple one-dimensional transport problem with the definitions of the Peclet and Courant numbers by

$$E \approx N_x \cdot N_t = \frac{L \cdot T \cdot w}{Pe^2 \cdot Co \cdot \delta^2 \cdot n \cdot R} \qquad (3)$$

where L is the length of the domain and T the overall time. It can be seen from equation (3) that the CPU-time required for a run is decreasing with the reverse of the squared Peclet number. Therefore an efficient algorithm has to overcome the Peclet number limit.

The flow and transport equation system represents a differential equation of composite type (parabolic - hyperbolic) and requires restrictions in space increment (grid Peclet number ≤ 2...10) and time step (COURANT-number ≤ 1). The enormous execution time requirement of solution methods for the mass transport equation results from the combination of these two drastic restrictions. Therefore, we developed the Front Limitation algorithm (Haefner *et al.*, 1996) that overcomes the Peclet condition. This way a grid can be taken as a basis which is fine to such an extent only as it is required by geological situation and hydrological boundary conditions regardless of the proportion of advective to dispersive transport within a cell of the grid (grid Peclet number). The essential characteristics of the FL- algorithm should be described briefly.

- The dispersive term of the mass transport equation is of second order and physically always leads to a concentration balance. Mathematically, this can be represented best by a cell-to-cell interpolation. The advective term is of first order and leads to a steep gradient. Therefore, it seems useful to extrapolate concentrations always from upstream to downstream and to obtain information about cell boundary concentration from this.
- The mass transport equation is solved in a semi-implicit way in order to obtain an accuracy of second order concerning time. An explicit treatment of the advective term is also possible and often recommendable, but it requires a relatively small Courant-number (< 0.25). The method has the lowest numerical dispersion (error) for the semi-implicit solution of dispersion and the explicit treatment of advection.
- In case of practical problems very different cell sizes are often required, especially very thin layers. These cells have an extremely low storage capacity. In such cells the steady state advective transport equation is only solved. This prevents oscillations in the concentrations

As a result a method was developed that is stable and sufficiently accurate for all grid Peclet numbers to simulate even pure advective processes (infinite Peclet number). The method was also included in our software package *CALIF* (*CALI*bration with *F*ront limitation) and in *VISUAL MODFLOW* (Waterloo Hydogeologic) and to execute on PC and workstations.

It should be mentioned that the numerical solution of a 2-dimensional transport problem with high advection (25×25 grid, Pe=100) has required a grid refinement to 250×250 cells ($Pe \leq 10$). Now the Front Limitation algorithm can do it with much coarser grids. Therefore it demands ca. one per cent of CPU-time. Such a drastic saving in CPU time is important for practical handling of reactive transport models.

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Nomenclature

c	volume concentration of a contaminant in the fluid [kg m^{-3}] .
Co	Courant number, $Co = \frac{w\,\Delta t}{n\,R\,\Delta x}$
C_{org}	content of organic carbon within sediment [we% C]
C_t	content of total carbon within sediment [we% C]
CEC	cation exchange capacity [meq/ 100g sediment]
$\boldsymbol{D}$	dispersion tensor
D	dispersion coefficient [m^2 s^{-1}] .
E	computer time expense
k	hydraulic conductivity [m s^{-1}] .
L	scale length [m] .
N_x, N_t	number of space and time increments
n	porosity [-] .
Pe	grid Peclet number, $Pe \approx \frac{\Delta x}{\delta_L}$
q	source strength of the transport equation [kg s^{-1} m^{-3}] .
R	retardation factor [-] .
S_{red}	content of reduced sulphur [we% S]
S_t	content of total sulphur [we% S]
t	time [s] .
T	overall time
TIC	content of total inorganic carbon within sediment [Ma% C]

w	Darcy velocity [m s^{-1}] .
x,y,z	coordinates [m] .
δ	dispersivity [m] .
λ	decay (degradation) coefficient [s^{-1}] .
Δx, Δt	cell increment, [m] and time increment (time step), [s].
Δz	layer thickness, [m]

Indices

k	species
L	longitudinal
t	transversal